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The Sieverts loop is an inexpensive, robust and reliable methodology for calculating hydrogen absorption in materials [1]. In this approach, we start by storing a sample of the material being tested in the volume V_{cell} (Figure 1) and initiate the process by producing a high vacuum in the system while the material sample is heated to eliminate (most of) the hydrogen and other impurities previously absorbed. The system typically operates isothermally, with the volume V_{ref} at ambient temperature and the sample at a temperature of interest – high enough to liquefy the alloy for the current application to nuclear fusion.

We then proceed with the absorption experiment by closing valve V_s , closing the valve leading to the vacuum pump, and opening the valve leading to the hydrogen source. A new pressure p_{ref} at temperature T_{ref} is established in the reference volume V_{ref} . Valve V_s is then opened and, after a suitable settling time, a new value of p_{sys} is measured once temperatures equilibrate at their desired (typically constant) values.

Measurement of hydrogen uptake is then made step-wise [2]. The valve V_s is closed after equilibration, and the reference volume V_{ref} is pressurized to sequentially larger pressures. Subsequent opening of the valve V_s allows equilibration at a new (higher) pressure p_{sys} . A mass balance enables calculation of absorbed hydrogen amounts.

Uncertainties in measurement of parameters, such as pressures, volumes, and temperatures, generate uncertainty in hydrogen absorption calculations, even when no measurement errors occur. It is therefore important to appropriately design the Sieverts loop to enable meaningful absorption measurements. This may be a challenge when the material being tested is a relatively weak absorbent of hydrogen. In our case, we start with lithium, which is a very good hydrogen absorbent. However, in the effort to reduce flammability, the lithium mole fraction may be reduced to the point where absorptivity drops below the capability of the Sieverts loop to accurately measure absorption. This may be the case even when proposed design rules are followed [3].

The purpose of this report is determining whether the Sieverts loop is capable of measuring hydrogen absorption at the conditions of interest for this project. In particular, we calculate hydrogen absorption and compare it with measurement uncertainty for two materials that approximately define the limits of interest for the project: pure lithium and eutectic lithium-lead (17% Li - 83% Pb mole fraction).

Analysis

The methodology presented here follows the approach outlined in [4] and calculates the amount of hydrogen absorbed based on a mass balance. The initial condition before step k in the pressurization process is a reference volume V_{ref} full of hydrogen to a specified pressure p_{ref}^k and the cell at a pressure p_{cell}^{k-1} . After valve V_s opening and equilibration, the whole system (reference and cell) ends up

at a uniform pressure p^k_{cell} . The mass absorbed during the k^{th} step can then be calculated by subtracting the mass in the gaseous phase before and after the opening of the valve,

$$m^k_{\text{abs}} = (m^{k-1}_{\text{ref}} + m^{k-1}_{\text{void}}) - (m^k_{\text{ref}} + m^k_{\text{void}}) \quad (1)$$

where m^k_{abs} is the mass absorbed in the k^{th} absorption step, and masses in the right hand side of the equation are in the gaseous (not absorbed) state. The gas masses in the reference volume and in the void space (cell volume minus sample volume) can be calculated with an appropriate equation of state for hydrogen.

Uncertainty can then be estimated from Equation 1 considering the uncertainty of individual variables (pressure, volume, temperature, hydrogen properties) [4]. The methodology has been programmed in an iterative equation solver, and was validated by comparison with examples given in [4]. The nature of the measurement in a Sieverts loop implies that the uncertainty in the absorbed mass will accumulate step by step. It is therefore important to demonstrate that the uncertainty accumulation remains a small fraction of the absorbed mass throughout the process.

Results

We have selected system dimensions and operating ranges appropriate for this project while following a series of design rules presented in a recent publication [3]. We have also estimated uncertainty in pressure, volume, and temperature measurements based on the instruments available for this project. System design parameters and measurement uncertainties are listed in Table 1.

We first consider lithium, and use absorption properties from [5]. As an excellent absorbent, 10 grams of lithium store almost 1 gram mole of hydrogen (Figure 2). With such high absorptivity, the measurement uncertainty (~ 0.001 mole after 20 absorption steps) is too small to be visible in the figure. We therefore anticipate being able to make meaningful measurements of lithium absorption – assuming no measurement errors.

For 17% lithium-83% lead molar mixtures, the results change. Lithium-lead [6] absorbs $\sim 100,000$ less hydrogen per unit of mass than lithium, and therefore uncertainty takes a much more important role. From Figure 3, measurement uncertainty has the same order of magnitude as hydrogen absorption. We therefore conclude that lithium-lead absorption cannot be meaningfully measured with the Sieverts loop as specified for this analysis.

Summary

We conclude that the Sieverts loop can produce meaningful results for lithium absorption with very small uncertainty. However, meaningful measurements of lithium-lead absorption and other poor hydrogen absorbents will require a different approach.

Future Work

With future funding, it would be possible to explore the possibility of using a differential Sieverts loop (Figure 4) to measure absorption into poorly absorbing alloys [7]. This system has several advantages: (1) pressure sensitivity is ~10 times higher because absorption measurement depends on differential pressure instead of total pressure, and (2) lower effect of temperature differences and hydrogen non-idealities because both sides experiment these effects equally. An uncertainty evaluation for this approach has been published [8] and could be applied to the problem of interest.

References

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Table 1. System design parameters and measurement uncertainties.

Parameter	Value	Uncertainty
Reference vessel volume	100 cm ³	0.005 cm ³
Cell vessel volume	100 cm ³	0.005 cm ³
Reference temperature	300 K	0.05 K
Cell temperature	600 K	0.05 K
Sample mass	10 g (Li), 100 g (LiPb)	0.01 g (Li), 0.1 g (LiPb)
Pressure	1-20 bar	600 Pa

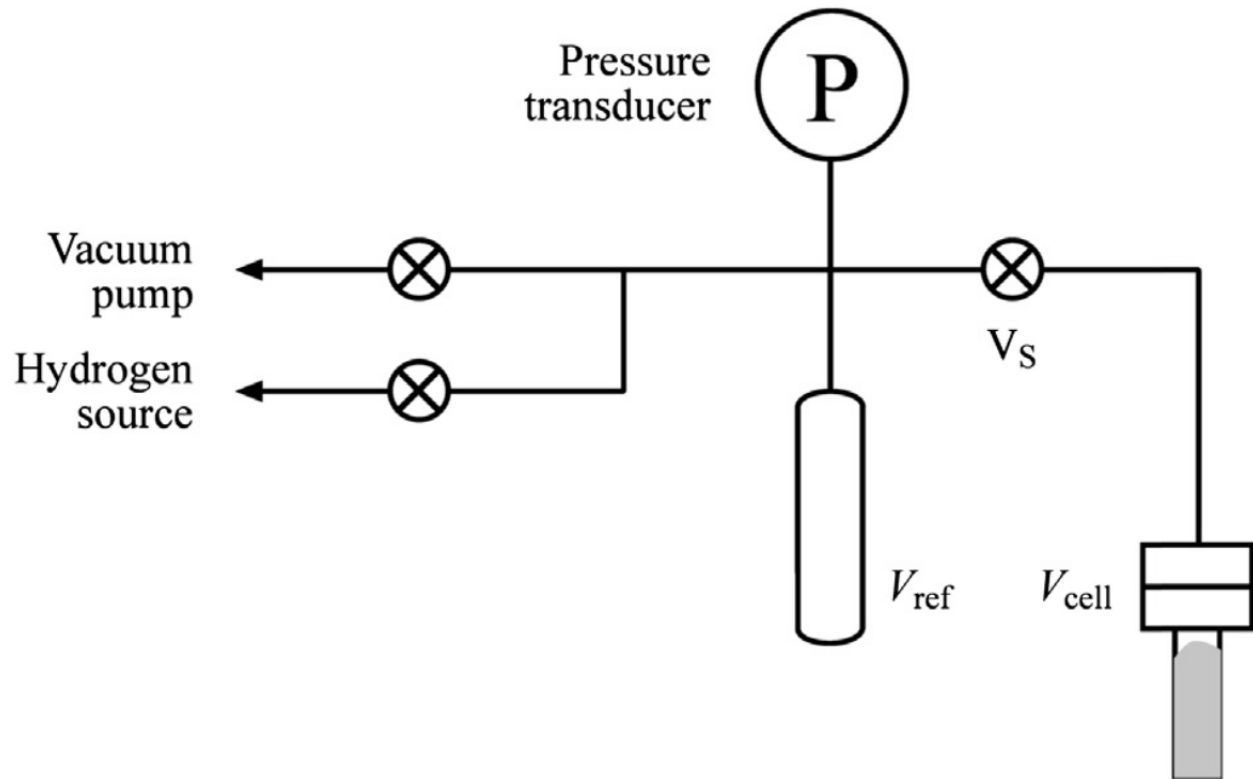


Figure 1. Sieverts loop for measuring hydrogen absorption in materials.

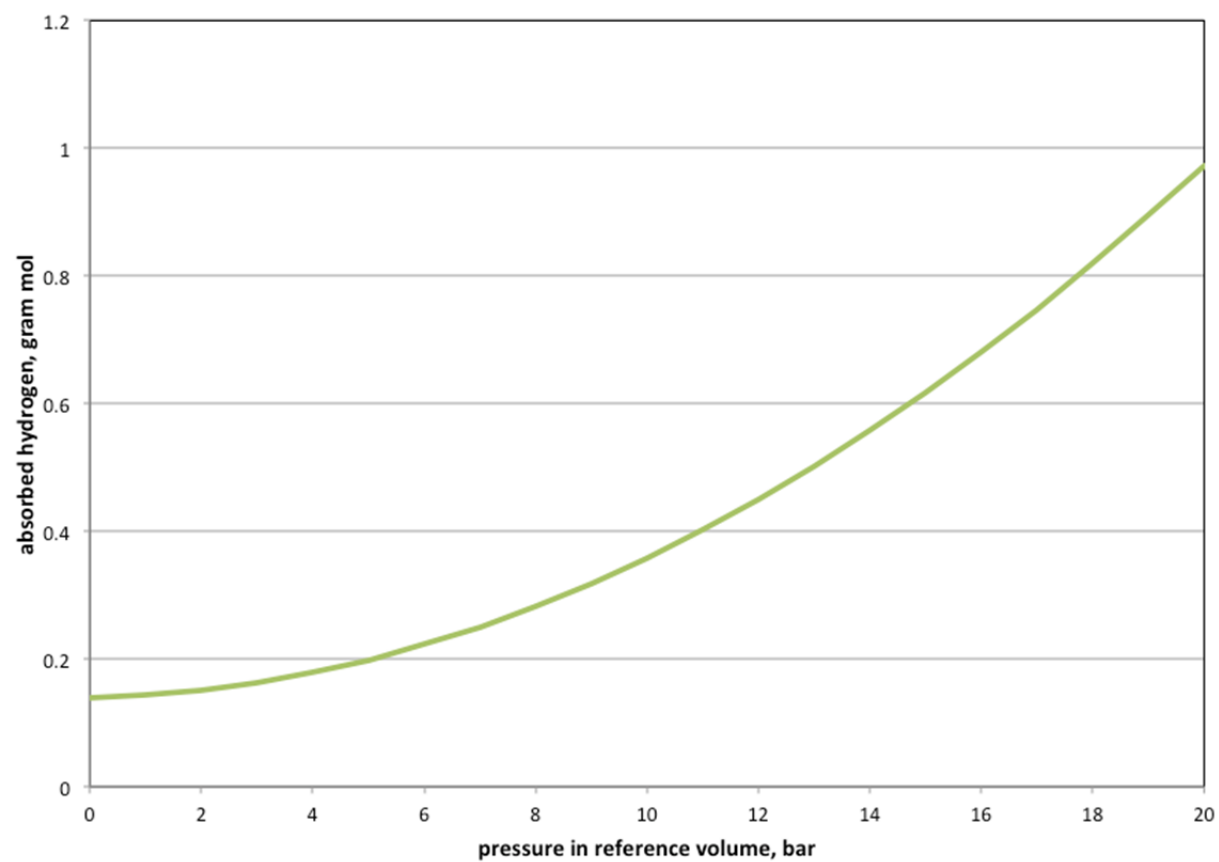


Figure 2. Number of gram moles of hydrogen absorbed in 10 g lithium sample.

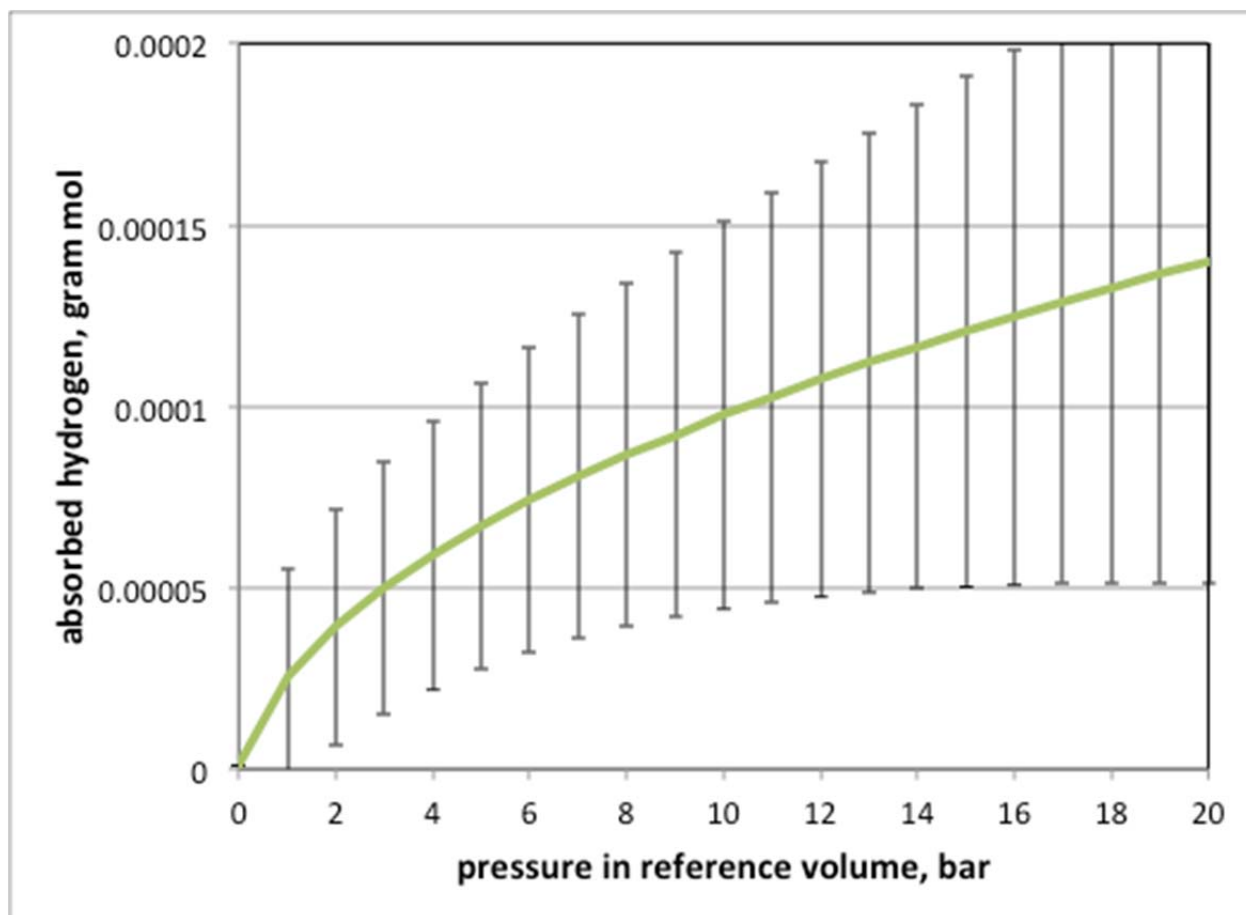


Figure 3. Hydrogen absorption in a 100 g eutectic lithium-lead mixture (17-83% mole fraction) with the measurement uncertainty in every step indicated by error bars.

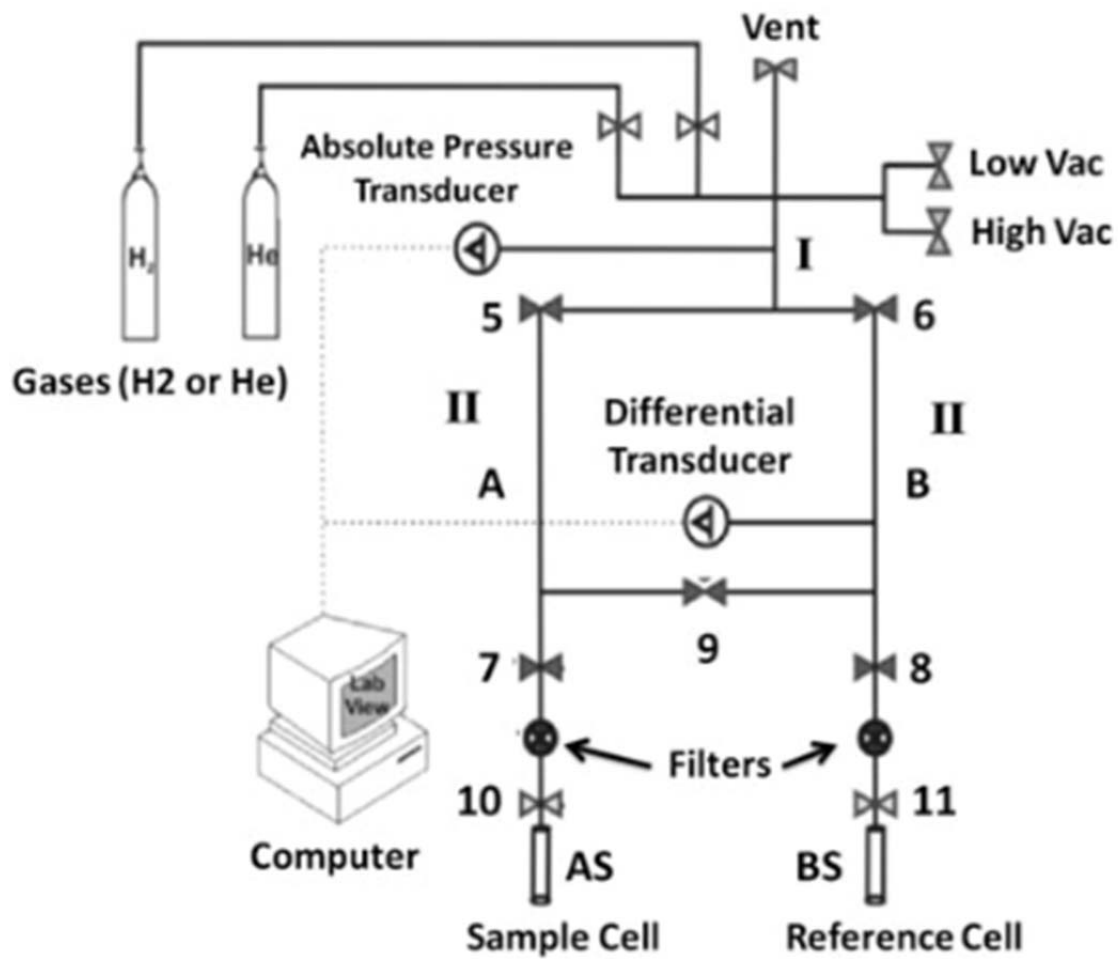


Figure 4. Proposed differential Sieverts loop for measurement of absorption characteristics of poorly absorbing alloys (Ref. 7).